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(54) METHOD FOR PRODUCING N-SUBSTITUTED IMIDAZOLE

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a method for producing an N-substituted imidazole and a method for producing a quaternary ammonium salt by using the same.

SOLUTION: A nitrile compound represented by the formula (1) $R_1CN...$ (1) (wherein R_1 is H, an aliphatic, aromatic or arylalkyl group) and an N-substituted 1,2-diamine represented by the formula (2) $H_2NCHR_2CHR_3NHR_4...$ (2) (wherein R_2 and R_3 are each independently H, an aliphatic, aromatic or arylalkyl group; R_4 is an aliphatic, aromatic or arylalkyl group) are allowed to react with each other in the presence of a metal oxide, a metal salt, an organic acid or a solid acid as a catalyst to form an N-substituted imidazoline. Then, the N-substituted imidazoline is dehydrogenated in the presence of one or more kinds of catalysts selected from the group consisting of nickel, palladium, platinum and copper.

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CLAIMS

[Claim(s)]

[Claim 1] The following general formula (1)

R1CN (1)

(R1 expresses the radical of a hydrogen atom or aliphatic series, aromatic series, or aroma aliphatic series among a formula.) The nitril compound shown and the following general formula (2)

H2NCHR2CHR3NHR4 (2)

(R2 and R3 express respectively independently the radical of a hydrogen atom or aliphatic series, aromatic series, or aroma aliphatic series among a formula.) R4 expresses the radical of aliphatic series, aromatic series, or aroma aliphatic series. As a catalyst the N-permutation 1 shown and 2-diamine A metallic oxide, The manufacture approach of N-permutation imidazole compound characterized by heating as a catalyst one sort chosen from the group which consists of nickel, palladium, platinum, and copper in N-permutation imidazoline compound which is made to react to the bottom of existence of a metal salt, an organic acid, or solid acid, and is obtained, or two sorts or more, and carrying out a dehydrogenation.

[Claim 2] The manufacture approach of N-permutation imidazole compound according to claim 1 that a substituent R1 is characterized by expressing the phenyl group permuted by a hydrogen atom, the alkyl group of carbon numbers 1-4, the phenyl group, the alkyl group of carbon numbers 1-4, or the alkoxy group in a general formula (1).

[Claim 3] In a general formula (2), substituents R2 and R3 become independent respectively. Hydrogen, the alkyl group of carbon numbers 1-4, The phenyl group permuted by the phenyl group, the alkyl group of carbon numbers 1-4, or the alkoxy group is expressed. The manufacture approach of N-permutation imidazole compound according to claim 1 or 2 that a substituent R4 is characterized by expressing the phenyl group permuted by the alkyl group of carbon numbers 1-4, the phenyl group, the alkyl group of carbon numbers 1-4, or the alkoxy group.

[Claim 4] The manufacture approach of N-permutation imidazole compound according to claim 1 to 3 characterized by carrying out continuously manufacture of N-permutation imidazoline compound, and manufacture of N-permutation imidazole compound under the same alcoholic existence.

[Claim 5] The manufacture approach according to claim 4 that alcohol is characterized by being fatty alcohol of carbon numbers 1-4.

[Claim 6] The manufacture approach of the 4th class imidazolium salt characterized by forming 4 class using the 4th class-ized agent which has a substituent which is different from N-substituent of this N-permutation imidazole compound in N-permutation imidazole compound obtained by the approach according to claim 1 to 5.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacture approach of N-permutation imidazole compound, and the manufacture approach of the 4th class imidazolium salt of having used it.

[0002] N-permutation imidazole compound can be used as an epoxy hardening accelerator. Moreover, by forming 4 class using the 4th class-ized agent which has a substituent which is different from N-substituent of this N-permutation imidazole compound in N-permutation imidazole compound, the 4th class imidazolium salt of asymmetry is obtained. The 4th class imidazolium salt of this asymmetry can be used as an electrolyte or an ionic liquid by selecting a specific anion kind.

[0003]

[Description of the Prior Art] Conventionally, the method of making alkyl halide react to the active hydrogen combined with the nitrogen atom in an imidazole as the manufacture approach of N-permutation imidazole compound is learned. However, by this approach, a reactant turns into mixture of N-permutation imidazole and an N and N'-2 permutation imidazole, and there is a problem that the yield of N-permutation imidazole falls.

[0004] As an approach which improved this, the approach to which an imidazole and alkyl halide are made to react under existence of an alkali is indicated by JP,43-12354,B. Or the attempt using carbonic acid diester with alkylation ability lower than ROGEN-ized alkyl also occurs. The method of making an imidazole and dimethyl carbonate react and specifically obtaining N-methyl imidazole is also proposed (refer to JP,9-169737,A).

[0005]

[Problem(s) to be Solved by the Invention] However, since a lot of by-products occur when an imidazole and alkyl halide are made to react under existence of an alkali, the excessive process which removes this is needed. On the other hand, although the approach of methylating using dimethyl carbonate can obtain N-methyl imidazole with comparatively high selectivity, when carrying out alkylation other than methylation, diethyl carbonate is expensive and it is difficult [it] to receive industrially.

[0006] This invention is made in view of the above-mentioned technical problem, and the purpose is offering the approach N-permutation imidazole compound's being easily manufactured by high yield.

[0007]

[Means for Solving the Problem] As a result of this invention persons' considering the manufacture approach of N-permutation imidazole compound wholeheartedly, N-permutation imidazole compound came to complete a header and this invention for being easily obtained by high yield by carrying out the dehydrogenation of the N-permutation imidazoline compound which a nitril compound, the N-permutation 1, and 2-diamine are made to react, and is obtained.

[0008] That is, this invention is the following general formula (1).

R1CN (1)

(-- R1 expresses the radical of a hydrogen atom or aliphatic series, aromatic series, or aroma aliphatic series among a formula.) -- the nitril compound shown and the following general formula (2)

H2NCHR2CHR3NHR4 (2)

(R2 and R3 express respectively independently the radical of a hydrogen atom or aliphatic series, aromatic series, or aroma aliphatic series among a formula.) R4 expresses the radical of aliphatic series, aromatic series, or aroma aliphatic series. As a catalyst the N-permutation 1 shown and 2-diamine A metallic oxide, It is the manufacture approach of N-permutation imidazole compound characterized by heating as a catalyst one sort chosen from the group which consists of nickel, palladium, platinum, and copper in N-permutation imidazoline

compound which is made to react to the bottom of existence of a metal salt, an organic acid, or solid acid, and is obtained, or two sorts or more, and carrying out a dehydrogenation.

[0009] This invention is further explained to a detail below.

[0010] The raw material compounds used in the approach of this invention are a nitril compound, the N-permutation 1, and 2-diamine.

[0011] In the approach of this invention, a nitril compound is a compound shown by the above-mentioned general formula (1), and its phenyl group permuted by a hydrogen atom, the alkyl group of carbon numbers 1-4, the phenyl group, the alkyl group of carbon numbers 1-4, or the alkoxy group is desirable as a substituent R1.

Although not limited especially as a nitril compound shown by the above-mentioned general formula (1), although hydrogen cyanide, an acetonitrile, propionitrile, butyronitrile, isobutyronitrile, phenylacetonitrile, torr nitril, a benzonitrile, a methyl benzonitrile, dimethylbenzo nitril, methoxybenzo nitril, dimethylbenzo nitril, naphth nitril, a cyano pyridine, etc. are illustrated, an acetonitrile, propionitrile, and butyronitrile are desirable from a reactant viewpoint in the ease of dealing with it, and, specifically, especially an acetonitrile is desirable.

[0012] Moreover, it sets to the approach of this invention and the N-permutation 1 and 2-diamine are the following general formula (2).

$H_2NCHR_2CHR_3NHR_4$ (2)

(-- R2 and R3 express respectively independently the radical of a hydrogen atom or aliphatic series, aromatic series, or aroma aliphatic series among a formula, and R4 expresses the radical of aliphatic series, aromatic series, or aroma aliphatic series.) -- it is the compound shown.

[0013] Although not limited in the approach of this invention especially as the N-permutation 1 shown by the above-mentioned general formula (2), and 2-diamine, as substituents R2 and R3 The phenyl group permuted respectively independently by hydrogen, the alkyl group of carbon numbers 1-4, the phenyl group, the alkyl group of carbon numbers 1-4, or the alkoxy group as a substituent R4 The phenyl group permuted by the alkyl group of carbon numbers 1-4, the phenyl group, the alkyl group of carbon numbers 1-4, or the alkoxy group is desirable. Specifically N-methyl ethylene diamine, N-ethyl ethylenediamine, N-propyl ethylenediamine, N-isopropyl ethylenediamine, N-butyl ethylenediamine, N-isobutyl ethylenediamine, N-benzyl ethylenediamine, N-phenylethylene diamine, N-methoxypheny ethylenediamine, N-dimethylphenyl ethylenediamine, N-tolyl ethylenediamine, the thing that transposed the ethylenediamine in the above-mentioned compound to 1,2-propanediamine further are illustrated. from a reactant viewpoint, N-methyl ethylene diamine, N-ethyl ethylenediamine, N-propyl ethylenediamine and N-methyl propanediamine, N-ethyl propanediamine, and N-propyl propanediamine are desirable, it is [among these] further alike, and they are N-methyl ethylene diamine and N-ethyl ethylenediamine preferably.

[0014] The N-permutation 1 first indicated to be the nitril compound shown by the above-mentioned general formula (1) by the above-mentioned general formula (2) and 2-diamine are made to react to the bottom of existence of a catalyst in the approach of this invention. Under the present circumstances, as a catalyst used, a metallic oxide, a metal salt, an organic acid, or solid acid can be used. As a metallic oxide, an aluminum oxide, a silicon dioxide, titanium oxide, a vanadium oxide, chromic oxide, manganese oxide, ferrous oxide, cobalt oxide, nickel oxide, oxidization copper, a zinc oxide, an oxidization yttrium, a zirconium dioxide, niobium oxide, molybdenum oxide, the silver oxide, cadmium oxide, indium oxide, oxidization tin, tantalum oxide, tungstic oxide, rhenium oxide, lead oxide, a lanthanum trioxide, cerium oxide, etc. are mentioned, for example. Moreover, as a metal salt, copper salt, zinc salt, iron salt, cobalt salt, manganese salt, an aluminum salt, tin<4> salt, mercury salt, chromium salt, and cadmium salt can be used. These salts may use any of organic-acid salts, such as inorganic-acid salts, such as a chloride salt, and acetate. Moreover, as an organic acid, methansulfonic acid, trifluoro methansulfonic acid, an ammonium chloride, an acetic acid, a phthalic acid, etc. are mentioned. Furthermore as solid acid, Nafion, a zeolite beta, etc. are mentioned. These catalysts may be used independently, and they may be used, mixing. The mixed catalyst of the viewpoint of catalytic activity to a methansulfonic acid, an ammonium chloride, zinc acetate or a zinc oxide, and an acetic acid is desirable among these catalysts.

[0015] In said approach of this invention, the amount of a nitril compound, the N-permutation 1, and 2-diamine used can react by one [the chemical equivalent or] excess.

[0016] In said approach of this invention, the amount of the catalyst used is usually used at a rate of 0.001-0.1 mols to the N-permutation 1 and one mol of 2-diamines.

[0017] In said approach of this invention, by making a metallic oxide, a metal salt, an organic acid, or solid acid

into a catalyst, a nitril compound, the N-permutation 1, and the reaction of 2-diamine are batch reactions, or can be performed by the successive reaction to the bottom of ordinary pressure or pressurization. At this reaction, in order that ammonia may generate during a reaction, reaction pressure rises, but this ammonia is also removable in the middle of a reaction, and after a reaction is completed, it is also removable. Moreover, a reaction can be performed in a 100 degrees C - 250 degrees C temperature requirement. At less than 100 degrees C, a reaction is so slow that it is not practical, above 250 degrees C, decomposition of amines arises and the yield of N-permutation imidazoline compound falls.

[0018] In said approach of this invention, although it is not necessary to use a solvent, since the viscosity of reaction mixture may rise when temperature is lowered, it is desirable to use a solvent. A solvent can be especially used without a limit, if inactive to a reaction condition. For example, although it is alcohols, the petroleum ether, benzene, toluene, etc., although water etc. disassembles an imidazoline compound, use's is not desirable. Alcohols with the effectiveness which activates the metal catalyst of this invention at the following dehydrogenation process among these solvents are desirable. That is, by using an alcoholic solvent, after manufacturing N-permutation imidazoline compound, dehydrogenation can be continuously performed efficiently by introducing the metal catalyst of this invention by reaction mixture as it is. As alcohols, from a viewpoint of solvent removal, on the fatty alcohol of carbon numbers 1-4, and a concrete target, methyl alcohol, ethyl alcohol, n-propyl alcohol, t-propyl alcohol, n-butyl alcohol, t-butyl alcohol, sec-butyl alcohol, etc. are desirable, and especially the low methanol of the boiling point is desirable.

[0019] It is desirable to react so that it may become the range of 0.1-10 by the weight ratio to N-permutation imidazoline compound obtained as an amount of solvents. Even if there is a possibility that the viscosity of imidazoline may rise and it exceeds 10, the effectiveness to which the solvent was made to increase is small at less than 0.1.

[0020] Even if it refines N-permutation imidazoline compound which is a resultant, it may be used for the following reaction process, without refining. Although various approaches, such as distillation and recrystallization, are learned, even if the purification approach of N-permutation imidazoline compound uses which approach, it does not interfere at all.

[0021] In the approach of this invention, dehydrogenation of the N-permutation imidazoline compound obtained at said process is carried out, and N-permutation imidazole compound is obtained. Under the present circumstances, it is necessary to heat them, using as a catalyst one sort chosen from the group which consists of nickel, palladium, platinum, and copper, or two sorts or more, and they need to carry out a dehydrogenation.

[0022] Since it is high activity as a catalyst, nickel is desirable and can use stabilization nickel, a Raney nickel catalyst, flake nickel, etc. as nickel further. According to a reaction format, that what is necessary is just to choose the optimal thing, the gestalt of a catalyst may be used with powder, and it may be used, fabricating.

[0023] In the approach of this invention, in case the dehydrogenation of the N-permutation imidazoline compound is carried out, as for the amount of catalysts used, it is desirable to react in 0.1 - 20% of the weight of the range to N-permutation imidazoline compound. Since a reaction rate becomes it slow that it is less than 0.1 % of the weight, disassembly of unreacted N-permutation imidazoline compound may arise, and the yield of N-permutation imidazole compound may fall. Even if it exceeds 20 % of the weight, the effectiveness to which the catalyst was made to increase is small.

[0024] In consideration of a by-product and coloring prevention, as for the dehydrogenation of this invention, it is desirable to carry out under the ambient atmosphere of inert gas, such as hydrogen, and nitrogen, helium, an argon, and before it reacts, it permutes a reactor by these gas, or it carries out aeration of these gas during a reaction, and is usually performed.

[0025] As for reaction temperature, in the dehydrogenation of this invention, it is desirable to carry out in 100 degrees C - 250 degrees C. Decomposition of a raw material and a product may arise at the temperature which is not practical since a reaction is very slow, and exceeds 250 degrees C at less than 100 degrees C. In order that hydrogen may generate this reaction during a reaction, reaction pressure rises. As for this hydrogen, it is desirable to remove, although it is not necessary to carry out even if it removes in the middle of a reaction.

[0026] In the dehydrogenation of this invention, it is desirable to perform a reaction under existence of a solvent. the alcohols which are the good solvents of N-permutation imidazoline industrially although there will be no limit especially if inactive [to a reaction condition] as a solvent -- desirable -- with a carbon number [the viewpoint of solvent removal to] of ten or less alcohol -- methyl alcohol, ethyl alcohol, n-propyl alcohol, t-propyl alcohol, n-butyl alcohol, t-butyl alcohol, sec-butyl alcohol, t-amyl alcohol, a cyclohexanol, 1-methyl

cyclohexanol, benzyl alcohol, etc. are specifically illustrated.

[0027] It is desirable to react in 0.1-10 by the weight ratio to N-permutation imidazoline compound as an amount of solvents in the dehydrogenation of this invention. Even if there is a possibility that imidazoline may join and it exceeds 10, the effectiveness to which the solvent was made to increase is small at less than 0.1.

[0028] In the approach of this invention, obtained N-permutation imidazole compound can be formed into 4 class using the 4th class-sized agent, and can manufacture the 4th class imidazolium salt.

[0029] N-substituent [in N-permutation imidazole compound obtained by the approach of this invention as the 4th class-sized agent in the approach of this invention -- the 4th class-sized agent which has a different substituent from substituent R4] in a general formula (2) -- it is necessary to use -- a carbon number -- 1-20 -- the thing of 1-12 can be used preferably. The following are mentioned as an example.

[0030] Dimethyl carbonate, methylethyl carbonate, diethyl carbonate, methyl carbonate-i-propyl, Carbonic acid di-i-propyl, methyl carbonate-n-propyl, carbonic acid G n-propyl, Carbonic acid di-n-butyl, carbonic acid G i-butyl, carbonic acid G t-butyl, carbonic acid G sec-butyl, Carbonic acid dipentyl, carbonic acid dihexyl, carbonic acid diheptyl, carbonic acid dioctyl, Carbonic acid dialkyls, such as carbonic acid dibenzyl, a dimethyl sulfate, a sulfuric-acid methylethyl, Diethyl sulfate, methyl sulfate-i-propyl, sulfuric-acid di-i-propyl, Methyl sulfate-n-propyl, sulfuric-acid G n-propyl, sulfuric-acid di-n-butyl, Sulfuric-acid G i-butyl, sulfuric-acid G t-butyl, sulfuric-acid G sec-butyl, Sulfuric-acid dipentyl, sulfuric-acid dihexyl, sulfuric-acid diheptyl, sulfuric-acid dioctyl, Sulfuric-acid dialkyls, such as sulfuric-acid dibenzyl, sulfurous-acid dimethyl, a sulfurous-acid methylethyl, Sulfurous-acid diethyl, methyl sulfite-i-propyl, sulfurous-acid di-i-propyl, Methyl sulfite-n-propyl, ***** G n-propyl, sulfurous-acid di-n-butyl, Sulfurous-acid G i-butyl, sulfurous-acid G t-butyl, sulfurous-acid G sec-butyl, Sulfurous-acid dipentyl, sulfurous-acid dihexyl, sulfurous-acid diheptyl, sulfurous-acid dioctyl, Sulfurous-acid dialkyls, such as sulfurous-acid dibenzyl, a methyl chloride, an ethyl chloride, Chlorination n-propyl, chlorination i-propyl, a n-butyl chloride, chlorination i-butyl, Chlorination t-butyl, chlorination sec-butyl, chlorination pentyl, chlorination hexyl, Chlorination heptyl, chlorination octyl, benzyl chlorides, and these chlorine A bromine, Alkyl halide, such as a thing replaced with iodine, phthalic-acid methyl, methyl benzoate, A methyl salicylate, methansulfonic acid methyl, trifluoro methansulfonic acid methyl, Trifluoroacetic acid methyl, p-toluenesulfonic-acid methyl, ethyl phthalate, Alkyl organic acids, such as ethyl benzoate, ethyl salicylate, methansulfonic acid ethyl, trifluoro methansulfonic acid ethyl, trifluoroacetic acid ethyl, and p-toluenesulfonic-acid ethyl, etc. can be illustrated.

[0031] It is carbonic acid dialkyls that it can exchange for a desired opposite anion easily after class[the / 4th]-izing among these, and there is no fear of impurity mixing, and especially its dimethyl carbonate is [among these] desirable. Moreover, sulfuric-acid dialkyls and alkyl halide are desirable from a viewpoint of a rate of reaction, and a dimethyl sulfate, a methyl chloride, and its methyl bromide are [among these] desirable. These can be properly used if needed from a viewpoint of an application and a manufacture process.

[0032] In the approach of this invention, although especially the mole ratio of N-permutation imidazole compound and the 4th class-sized agent does not have a limit, in order to complete a reaction, the 4th class-sized agent of the way of an excessive amount is good. Usually, the N-permutation imidazole compound:4th class-sized agent = it is 1:3-1:1 and is 1:2-1:1 preferably.

[0033] or [not reacting to the 4th class-sized agent in the approach of this invention as the solvent by the reaction of N-permutation imidazole compound and the 4th class-sized agent using a solvent] -- or it is good the anything which cannot react easily. The organic solvent which generally has a polarity from a viewpoint of a reaction rate is desirable. Specifically Methyl alcohol, ethyl alcohol, n-propyl alcohol, i-propyl alcohol, n-butyl alcohol, i-butyl alcohol, sec-butyl alcohol, T-butyl alcohol, a cyclohexanol, benzyl alcohol, Ethylene glycol, propylene glycol, a diethylene glycol, Alcohols, such as triethylene glycol and a glycerol, an acetonitrile, Propionitrile, butyronitrile, valeronitrile, a benzonitrile, Ketones, such as nitril, such as acrylonitrile, an acetone, and a methyl ethyl ketone Sulfoxides, such as dimethyl sulfoxide, a sulfolane, and 3-methyl sulfolane A formamide, N-methyl formamide, N,N-dimethylformamide, Heterocycle type solvents, such as lactone, such as amides, such as an acetamide, N-methyl acetamide, N,N-dimethylacetamide, and N-methyl pyrrolidone, and other gamma-butyrolactone, and 1,3-dimethyl-2-imidazolidinone, etc. can be illustrated. Although the optimal solvent changes by the 4th class-sized agent, generally a methanol and an acetonitrile are desirable.

[0034] Although it is not limited, when a solvent is used for it, and a reaction rate and the removal nature of heat of reaction are taken into consideration, as for especially the amount of the solvent used in the reaction of N-permutation imidazole compound and the 4th class-sized agent, it is desirable to make it 10 or less times by

the weight ratio to N-permutation imidazole compound. Even if it exceeds 10, the effectiveness which increased the solvent is small.

[0035] It is more desirable to carry out by pressurization-ization, when using the 4th class-sized agent of a low-boiling point although the reaction of N-permutation imidazole compound and the 4th class-sized agent may be performed under ordinary pressure or you may carry out under pressurization.

[0036] In the reaction of N-permutation imidazole compound and the 4th class-sized agent, although it is dependent on the boiling point of the 4th class-sized agent and an organic solvent when carrying out by ordinary pressure, 40-250 degrees C of reaction temperature are usually 50-200 degrees C preferably.

[0037] Generally the raw material compound and solvent which are used in the approach of this invention are marketed, and the approach of this invention can be enforced using them.

[0038]

[Example] Hereafter, although an example explains the approach of this invention, this invention is not limited to these. In addition, a gas chromatography (Shimadzu make) and high performance chromatography (TOSOH CORP. make) analyzed the purity analysis in an example.

[0039] After putting in and carrying out the nitrogen purge of N-ethyl ethylenediamine 61.7g, acetonitrile 31.6g, methanol 40.0g, and the 7.0g of the zinc acetate to the autoclave made from 1200ml stainless steel of examples, it heated at 200 degrees C and reacted for 3 hours. When this was cooled after reaction termination and gas chromatography analyzed, N-ethyl ethylenediamine invert ratio was 93%, and the selectivity of 1-ethyl-2-methyl imidazoline was 99%.

[0040] After it put stabilization nickel (JGC chemistry company make, N103B) 3.2g into the 200ml autoclave made from stainless steel and it carried out the nitrogen purge to the obtained 1-ethyl-2-methyl imidazoline reaction solution, it heated at 180 degrees C and the reaction was performed for 5 hours. The hydrogen to generate was extracted through piping with a bore of 1mm during the reaction. When it cooled to the room temperature after reaction termination and gas chromatography analyzed, 90% of invert ratios of 1-ethyl-2-methyl imidazoline and the selectivity of 1-ethyl-2-methylimidazole were 92%.

[0041] After putting in and carrying out the nitrogen purge of N-ethyl ethylenediamine 61.7g, acetonitrile 31.6g, methanol 40.0g, and the 3.2g of the copper(II) oxide to the autoclave made from 1200ml stainless steel of examples of reference, it heated at 200 degrees C and reacted for 3 hours. When this was cooled after reaction termination and gas chromatography analyzed, N-ethyl ethylenediamine invert ratio was 70%, and the selectivity of 1-ethyl-2-methyl imidazoline was 98%.

[0042] After putting in and carrying out the nitrogen purge of N-ethyl ethylenediamine 61.7g, acetonitrile 31.6g, methanol 40.0g, and the 3.7g of the methansulfonic acid to the autoclave made from 2200ml stainless steel of examples of reference, it heated at 180 degrees C and reacted for 5 hours. When this was cooled after reaction termination and gas chromatography analyzed, N-ethyl ethylenediamine invert ratio was 91%, and the selectivity of 1-ethyl-2-methyl imidazoline was 98%.

[0043] After it put 1-ethyl-2-methylimidazole 33.0g [which was obtained in the example 2 example 1], 54.0g [of dimethyl carbonate], and methanol 50g into the 200ml autoclave made from stainless steel and it carried out the nitrogen purge, it heated at 135 degrees C and the reaction was performed for 7 hours. The invert ratio to 1-ethyl -1 and 2-dimethyl imidazolium monomethyl carbonate was 94.0%.

[0044] Having taught 1-ethyl-2-methylimidazole 33.0g and acetonitrile 100g compounded by the same approach as example 3 example 1 to the 200ml autoclave made from stainless steel, and supplying 16.0g of methyl chlorides to it, it heated at 50 degrees C and the reaction was performed for 5 hours. The invert ratio to 1-ethyl -1 and 2-dimethyl imidazolium chloride salt was 97.0%.

[0045] 2-methylimidazole 24.6g and acetonitrile 75.0g were taught to the autoclave made from 1200ml stainless steel of examples of a comparison, and the reaction was performed at 70 degrees C for 4 hours, supplying 20.3g of ethyl chlorides to it. When this was cooled and the high speed liquid chromatography analyzed the reactant, they were 40 mol % of 1, 2, and 3-trimethyl imidazolium chloride salts, and 2-methylimidazole 10 mol % 1-ethyl-2-methylimidazole 50 mol%.

[0046] It reacted by the completely same approach as the example 1 of a comparison except having added 12.0g of example of comparison 2 sodium hydroxides. When the high speed liquid chromatography analyzed the reactant, they were five mol % of 1, 2, and 3-trimethyl imidazolium chloride salts, and 2-methylimidazole 5 mol % 1-ethyl-2-methylimidazole 90 mol%. Moreover, the sodium chloride carried out about 16g byproduction.

[0047]

[Effect of the Invention] According to the approach of this invention, N-permutation imidazole compound used as an epoxy hardening accelerator can be easily manufactured by high yield, and the approach of this invention is very useful industrially.

[0048] Moreover, N-permutation imidazole compound obtained by the approach of this invention can obtain the 4th class imidazolium salt of asymmetry by forming 4 class using the 4th class-ized agent which has a different substituent from the N-substituent. By selecting a specific anion kind, the 4th class imidazolium salt of asymmetry can be used as an electrolyte or an ionic liquid, and is very useful industrially.

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